Landfill Leachate Treatment by Aerated Recirculation and Pressurized Suspended Fiber Biofiltration

Boya Wang[1], Peter Grasel[2], Gary Millington[3], John Hallas[4], Hafiz Ahmad[5], Gang Chen[6*]

[1,6]Department of Civil and Environmental Engineering, FAMU-FSU College of Engineering, Tallahassee, FL 32310

[2,3]Florida Department of Environmental Protection, Tallahassee, Florida 32399

[4]Talquin Electric Company, Tallahassee, Florida 32399

[5]Florida State University Panama City Campus, Panama City, Florida 32405

[1]bw14c@my.fsu.edu; [2]Peter.Grasel@dep.state.fl.us; [3]Gary.Millington@dep.state.fl.us; [4]jhallas@talquinelectric.com; [5]hahmad@pc.fsu.edu; [6]gchen@eng.fsu.edu

Abstract

Recirculating leachate appears to be one of the least expensive methods for partial treatment and disposal of leachate at properly designed and operated landfill sites. The leachate recirculation not only improves the leachate quality, but also shortens the time duration required for landfill stabilization. In addition, aerated leachate recirculation may bring air into the landfill, leading to aerobic degradation of organic compounds and precipitation of heavy metals. Although aerated leachate recirculation has these advantages, leachate is only partially treated. The leachate must be further treated in order to meet the discharge requirements. In this research, aerated recirculation and pressurized suspended fiber biofiltration were tested for the treatment of leachate from landfills in Northwest Florida. The pressurized suspended fiber biofilter was configured for biological contact oxidation, a novel and efficient treatment process for enhanced organic and iron removal. Using the combined aerated leachate recirculation and pressurized fiber biofiltration, chemical oxygen demand (COD) of the landfill leachate can be reduced to as low as 8 mg/l and iron content can be reduced to as low as 0.02 mg/l under appropriate dissolved oxygen and alkalinity conditions.

Keywords

Aerated Recirculation; Suspended Fiber; Biofiltration; Landfill Leachate and Iron

Introduction

Landfilling is widely adopted as one of the most economical processes for solid waste disposal. At the same time, landfill leachate is of great environmental concern because of its complex composition and high contaminant concentrations (Ragle et al., 1995). To prevent/reduce pollution of the natural environment, technologies including biological degradation, chemical and electrochemical oxidation, coagulation-flocculation, precipitation, adsorption, ion exchange and reverse osmosis, etc. have been practiced for the treatment of landfill leachate (Coban et al., 2012; Renou et al., 2008; Wintheiser, 1998). Currently, as one of the least expensive methods for partial treatment and disposal, leachate recirculation has drawn more and more attention, especially at properly designed and operated landfill sites (Reinhart and AlYousfi, 1996; Yang et al., 2012). Through leachate recirculation, organic contents and heavy metals can be significantly reduced. The leachate recirculation not only improves the leachate quality, but also shortens the time duration required for landfill stabilization.

The biological reactions during leachate recirculation are basically anaerobic. However, aerobic conditions may prevail if the recirculated leachate is aerated. Recently, increased interest has been focused on introducing air into the waste mass for aerobic degradation of solid waste since aerobic processes have been promoted as a method for accelerating solid waste stabilization. Studies of aerobic biodegradation processes have demonstrated that the organic parts of the refuse can be degraded in a relatively shorter time when compared with those of anaerobic degradation processes (Bilgili et al., 2007; Nikolaou et al., 2010). The concept of aerobic degradation by introducing air into a landfill presents significant alternatives in waste management both for existing and new systems. In

2326-5787/15/01 007-12 © 2015 DEStech Publications, Inc.
doi: 10.12783/fiee.2015.0401.02

addition to promotion of aerobic organic decomposition, aerated recirculation can also help iron precipitation as well as nitrogen nitrification. In Northwest Florida, high concentrations of iron have been observed in the landfill leachate, which is believed to be released to the landfill leachate from the iron-rich soil owing to changes in pH and redox conditions induced by organic waste decomposition. Therefore, aerated leachate recirculation has obvious benefits in this region.

Although aerated leachate recirculation has above advantages, leachate is only partially treated through aerobic recirculation. The leachate must be further treated in order to meet the discharge requirements. Especially, since leachate has been partially treated after aerated recirculation, more efficient treatment methods are required to further treat the leachate. Biological treatment of wastewater with a biofilter is among the oldest and most well characterized technologies (Ferraz et al., 2014). During biofiltration operations, the growth of microorganisms develops biofilms on the medium surfaces and the microorganisms in the biofilms absorb soluble and colloidal waste materials in the wastewater as it percolates over the medium surfaces. Recently, polypropylene fibers have been introduced as the biofilter media. Subsequently, pressurized suspended fiber biofilters have been practiced in drinking water and wastewater treatment as a space-saving technology (Lee et al., 2008). The pressurized suspended filter pore space (and subsequent retention time) can be adjusted, which offers the flexibility to achieve different filtration functions. In addition, pressurized suspended fiber biofilters have other obvious advantages, the most important one of which is that the suspended fibers provide a tremendous amount of surface areas in a small volume. Therefore, microorganisms can grow on the fibers at a density greater than 1×10^8 cells per ml, the only means to culture cells at in vivo-like cell density (Chaiprasert et al., 2003). Another advantage of the pressurized suspended fiber biofiltration is that the oxygen transfer barrier can be overcome and significantly increased dissolved oxygen level can be achieved. Prior studies have demonstrated that BOD and COD removal increases with the increase of pressure when the pressure is raised up to 6 bars in a laboratory scale rotating biological contactor (Ellis et al., 1992). As an innovative technology, the pressurized suspended fiber biofilters also makes biological contact oxidation possible, which can significantly improve organic removal and decrease the sludge yield. For iron removal, contact oxidation is achieved by microbial mediated iron oxidation and fixation during which ferrous iron is oxidized to ferric iron and fixed onto the filter media. There is minimal ferric iron suspending in the solution that can escape the filter.

In this research, leachate collected from landfills in Northwest Florida was treated by aerated recirculation and suspended fiber biofiltration in laboratory bioreactors. For aerated recirculation, variable recirculation cycles as well as aeration levels were investigated. The pressurized suspended fiber biofilter was designed and operated to achieve biological contact oxidation, which removed organic compounds and iron more efficiently. The pressurized suspended fiber biofiltration, a cost- and space-saving technology, combined with aerated leachate recirculation, provided a new alternative means for the treatment of landfill leachate.

Material and Methods

Leachate, Solid Waste and Soil Collection and Characterization

Landfill leachate and solid waste were collected from the Leon County Landfill, located in Tallahassee, FL and Springhill Landfill, located in Campbellton, FL. Leon County Landfill accepts class III commercial and residential waste through Marpan Recycling, which includes yard trash, C&D debris, processed tires, asbestos, carpet, cardboard, paper, glass, plastic, furniture other than appliances, and other materials approved by Florida Department of Environmental Protection. Besides domestic waste from Leon County, Springhill Landfill also accepts other non-hazardous waste including auto shredder fluff, biosolid, drum management-liquid, drum management-solid, and liquifix (by solidification), etc. The leachate was collected from these two landfills in temperature-controlled containers at 4^{o}C and transported to the laboratory immediately and stored under refrigeration at 4^{o}C until the reaction. The collected leachate mainly included four groups of pollutants, i.e., dissolved organic matter, inorganic macro-components, heavy metals, and xenobiotic organic compounds. The dissolved organic matter was composed of fulvic-like and humic-like compounds; inorganic macro-components were composed of calcium (Ca^{2+}), magnesium (Mg^{2+}), sodium (Na^+), potassium (K^+), ammonium (NH_4^+), iron (Fe^{2+}), manganese (Mn^{2+}), chloride (Cl^-), sulfate (SO_4^{2-}) and hydrogen carbonate (HCO_3^-); heavy metals were composed of

cadmium (Cd^{2+}), chromium (Cr^{3+}), copper (Cu^{2+}), lead (Pb^{2+}), nickel (Ni^{2+}) and zinc (Zn^{2+}); and xenobiotic organic compounds were originated from household or industrial chemicals and present in relatively low concentrations, which were composed of variety of aromatic hydrocarbons, phenols, chlorinated aliphatics, pesticides, and plastizers, etc. The major concerns of the leachate from these two landfills were dissolved organic matter which was quantified by chemical oxygen demand (COD) and iron that was released to the landfill leachate from the iron-rich soil. The Floridian soil is mainly composed of Myakka, an acid soil characterized by a subsurface accumulation of humus and Al(III) and Fe(III) oxides. In Northwest Florida, iron content is much higher than the rest of the state with a range of 20 to 200 mg/g soil (Williams et al., 2011). In the subsurface soil, iron exists in the form of Fe(III) oxides. When reducing conditions occur, iron reducing bacteria can reduce various forms of Fe(III)-oxides and generate and release large concentrations of soluble Fe(II).

The solid waste was also collected from these two landfills. The collected solid waste was ground and sieved before introduced into the laboratory bioreactors. Besides leachate and waste, soil samples were collected from the landfill site and used as inocula for microbial cultivation and iron content assessment. To assess the soil iron content, soil samples were first partially thawed and placed in an anaerobic chamber with a maintained H_2-N_2 atmosphere. The samples were then ground and the weighed samples were placed in a glass reaction vessel and purged with CO_2-scrubbed air, after which the samples were acidified with hot, 5% perchloric acid to dissolve carbonate precipitates such as siderite, calcite, aragonite, and carbonate forms of green-rust. Evolved CO_2 gas was carried to the coulometer cell containing a CO_2-sensitive ethanolamine solution and quantitatively titrated. The samples were then reacted with 0.25 M hydroxylamine (NH_2OH) hydrochloride in 0.25 N HCl and incubated at 60°C for 2 hrs for iron extraction (Lovley and Phillips, 1988; Roden and Zachara, 1996). Following the extraction, soil iron content was determined using a spectrophotometer at the wavelength of 447 nm (Shimadzu UV-1650 PC).

Aerated Leachate Recirculation Investigation

A laboratory scale recirculation bioreactor was set up to simulate landfill leachate recirculation, which was followed by a pressurized suspended fiber biofilter. The custom-made recirculation bioreactor had a working volume of 35 liters with a height to diameter ratio of 1:1.66 (height = 50 cm and diameter = 30 cm). The reactor was packed with solid waste that was collected from above-mentioned landfills. To avoid the void resulted by large solid waste that may lead to flow shortcut in the bioreactor, the solid waste was ground and sieved (< 2 mm). In real case scenarios, since the typical depth of a landfill is around 20 meters, recirculated leachate has a good chance to interact with the solid waste as simulated in above design. Within the bioreactor, a CO_2 entrapment device was arranged. The consortia that were responsible for organic degradation were cultured first and then introduced to the bioreactor. The consortia were cultured using the sampled soil as the innocula under conditions of the bioreactor with collected leachate as the carbon and energy sources for organic decomposition. After aeration, the collected landfill leachate was pumped to the bioreactor from the storage reservoir using a peristaltic pump at a flow rate of 100 ml/min. Leachate aeration was achieved in the storage reservoir with targeted dissolved oxygen levels of 3 mg/l to 6 mg/l as measured directly by a dissolved oxygen meter. Before being introduced to the bioreactor, the pH of the leachate was adjusted with 1 M NaOH to 7.5 and 8.5, respectively since obvious iron removal can only be achieved with pH greater than 7.5. Aerated leachate recirculation was investigated for 10 recirculation cycles. After each cycle, leachate was monitored for chemical oxygen demand (COD) and total iron by spectrophotometric analysis techniques using a spectrophotometer at the wavelength of 447 nm (Shimadzu UV-1650 PC).

Pressurized Suspended Fiber Biofiltration Performance Evaluation

A custom-made pressurized suspended fiber biofilter with a working volume of 35 liters was used for this part of research. This reactor had a height to diameter ratio of 1:1.66 (height = 50 cm and diameter = 30 cm). In the pressurized suspended fiber biofilter, polypropylene fibers were arranged to be suspended in the bioreactor. Two water rubber bags were arranged on two sides of the reactor, each with a capacity of 8 liters (Figure 1). When the water bags were filled with water, the suspended fibers were squeezed together, producing minimal pore space for enhanced filtration. With the ongoing filtration, microbial growth and metal precipitation, the pore space was occupied and pressure was built up. Consequently, water was withdrawn from the water bags to release filter pore space for further filtration. When the water bags were fully filled, the working volume of the pressurized

suspended fiber biofilter was a half of the volume of the bioreactor. In the pressurized suspended fiber biofilter, organic compounds were continued to be decomposed by inoculated consortia similar as those in the recirculation bioreactor. These consortia strategically positioned themselves on the suspended fiber to form a biofilm, which had demonstrated to be able to degrade complex molecules such proteins, carbohydrates and lipids. Iron removal, especially ferrous iron that was released at the bottom of the recirculation bioreactor when oxygen was depleted can be removed through contact oxidation, during which iron oxidation and fixation occurred at the same time.

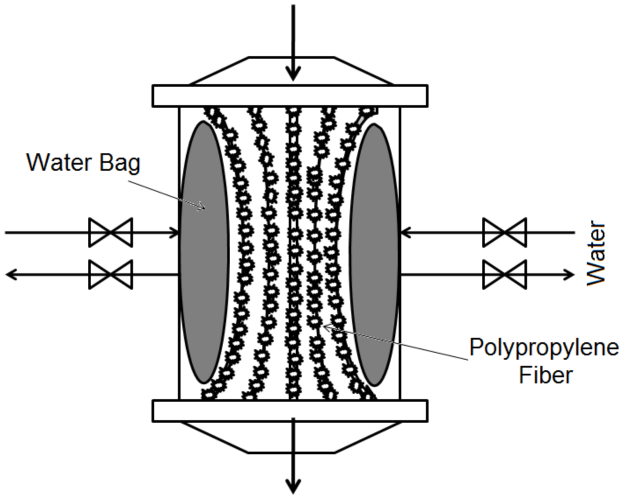

FIGURE 1. ILLUSTRATION OF PRESSURIZED SUSPENDED FIBER BIOFILTER. SUSPENDED FIBERS WERE SQUEEZED TOGETHER AT THE BEGINNING OF FILTRATION, PRODUCING MINIMAL PORE SPACE. WITH THE ONGOING FILTRATION, MICROBIAL GROWTH AND METAL PRECIPITATION, PORE SPACE WAS OCCUPIED AND PRESSURE WAS BUILT UP. SUBSEQUENTLY, WATER WAS WITHDRAWN FROM THE WATER BAGS TO RELEASE FILTER PORE SPACE FOR FURTHER FILTRATION.

During the operation, the pressure was maintained at 3 bars. The water bags were first filled with water and compressed air was supplied to the biofilter to achieve dissolved oxygen levels of 1 mg/l, 2 mg/l and 3 mg/l, respectively, which was controlled by a mass-flow controller as monitored by a dissolved oxygen meter. Leachate was introduced to the biofilter using a peristaltic pump at a flow rate of 50 ml/min. With the ongoing of the biofiltration and building up of the pressure, water was withdrawn decrementally from the water bags to release the pore space and reduce the pressure. Organic decomposition and iron oxidization as a function of alkalinity was also investigated. Specifically, at dissolved oxygen level of 2 mg/l, alkalinity was adjusted with lime up to 800 mg/l as $CaCO_3$ before the leachate was introduced to the pressurized suspended fiber biofilter.

For the convenience of this research, a batch-based instead of continuous leachate treatment was conducted. Therefore, the recirculation bioreactor and pressurized suspended fiber biofilter had the same size. For practical applications, a continuous treatment requires proper sizing of the corresponding bioreactors.

Model Development

Iron removal and release in the recirculation bioreactor can be described by the expression with iron removal following a first order reaction and iron release following an exponential function:

$$\frac{dC}{dt} = -k_1 \cdot C + a \cdot e^{-k_2 C} \qquad \text{Equation (1)}$$

where k_1 is the iron removal rate coefficient and k_2 is the iron release rate coefficient, respectively. In above equation, iron release and removal are described in two separate terms, which should well describe the fate of iron in the recirculation bioreactor.

Iron and organic removal in the pressurized fiber biofilter was quantified in terms of removal coefficient, k_c:

$$k_c = -\frac{v}{L} Ln(1 - fr) \qquad \text{Equation (2)}$$

where k_c is the removal coefficient; L is the length of the bioreactor; v is the velocity; and fr is the percentage removal.

Theoretically, the energy that microorganisms obtained from the organic oxidation or iron oxidation must balance their need to synthesize the new cells (McCarty, 1975). Consequently,

$$\varepsilon A \Delta G_r + \Delta G_s = 0 \qquad \text{Equation (3)}$$

where ε is the efficiency of energy transfer to or from the energy carrier (e.g., ATP) which is assumed to be 0.6; ΔG_r is the free energy released per electron equivalent (eeq) (amount of the substrate that releases 1 mole e^- during a specified oxidation reaction) of electron-donor substrate converted for energy (e.g., respiration); ΔG_s is the carrier (ATP) energy required to synthesize 1 eeq of cells which includes energy loss incurred in using the energy carrier (e.g., ATP); and A is the balance ratio between ΔG_r and ΔG_s. For heterotrophic growth with ammonia as nitrogen source, A can be estimated by (McCarty, 1975):

$$A = \frac{\frac{-\Delta G_p}{\varepsilon^m} - \Delta G_c}{\varepsilon \Delta G_r} \qquad \text{Equation (4)}$$

where ΔG_p is the free energy required (or evolved) in conversion of the carbon source to pyruvate (kcal per eeq pyruvate); ΔG_c is the ATP energy required to form 1 eeq cells from pyruvate and ammonia which is assumed to be 7.5 kcal; m = +1 when $\Delta G_p > 0$ and m = -1 when $\Delta G_P < 0$. For autotrophic growth with ammonia as the nitrogen source, A is estimated by:

$$A = \frac{-59.7}{\varepsilon \Delta G_r} \qquad \text{Equation (5)}$$

Stoichiometric yield coefficient Y, i.e., biomass formed per unit of substrate consumed, can be estimate as:

$$Y = \frac{\alpha}{\beta(1+A)} \qquad \text{Equation (6)}$$

where α is the mole weight of 1 eeq biomass which equals to 5.65 g for ammonia served as the nitrogen source if bacteria are assumed to have a formula of $C_5H_7O_2N$ and β is the molar weight of 1 eeq substrate which equals to 7.5 g, 5.33 g and 2.78 g for carbohydrates, proteins and fat, respectively.

Results

Soil and Leachate Characterization

The soil samples were characterized based on sieve analysis and were identified as loamy or fine sand for Leon County Landfill and Springhill Landfills. Based on sieve analysis, all the soil samples exhibited a poor grading, i.e., the soil particles were in general similar in size range. The finest particles were screened out by sieve 200 (~ 75μm). Leon County Landfill soil had a percentage fine of 5.22% and Springhill Landfill soil had a percentage fine of 4.55%. The soil iron content was 43.8 mg/g for Leon County Landfill soil and 34.0 mg/g for Springhill Landfill soil. It should be noted that only reducible iron contributed to the quantified iron content. There was a general trend that the soil iron content increased with the increase of percentage of finer particles. This is due to the increase in surface area available for iron accumulation. The leachate characterization indicated that the Leon County Landfill leachate had a composition of COD of 9872 mg/l, BOD_5 1184 mg/l, NH_4^+ of 274 mg/l, NO_3^{-1} of 31.8 mg/l and pH of 7.02. On the other hand, Springhill Landfill leachate had a composition of COD of 13887 mg/l, BOD_5 of 2023 mg/l, NH_4^+ of 509 mg/l, NO_3^{-1} of 61.4 mg/l and pH of 7.42.

Aerated Recirculation

At pH 7.5, dissolved oxygen played an important role in iron oxidation and removal. At this pH level, it seemed that dissolved oxygen of 5 mg/l and above would ensure iron oxidation and removal (Figure 2). At pH 8.5, further increase of dissolved oxygen above 3.0 mg/l did not make significant difference in iron removal. Iron removal became stable after 4 recirculation cycles and above 98% of iron was removed for both Leon County Landfill leachate and Springhill Landfill leachate. High iron removal at high pH was owing to the fact that iron hydroxide speciation was a function of pH. At pH 7.5, around 50% of iron would form iron hydroxide. On the other hand, above 95% iron would precipitate in the form of iron hydroxide at pH 8.5. The experimental results demonstrated

that high dissolved oxygen and pH ensured high levels of iron existing in the form of iron hydroxide, which could be consequently removed from landfill leachate during recirculation by precipitation. Iron concentration was simulated against Equation (1) by means of non-linear regression of simplex optimization of least squares. 95% confidence intervals and prognosis intervals were determined for each fitted curve. The simulated iron removal rate coefficient was in the range of 0.6 hr^{-1} to 0.8 hr^{-1} and the iron release rate coefficient was in the range of 0.4 hr^{-1} to 2.6 hr^{-1}. Leon County Landfill had greater iron removal rate coefficient and iron release rate coefficient than those of Springhill Landfill (Figure 3). For both landfills, the iron removal rate coefficient was higher at pH 8.5 than that of pH 7.5, which increased with the increase of dissolved oxygen. The iron release rate coefficient decreased with the increase of dissolved oxygen, which was more pronounced at pH 7.5 than that of pH 8.5 (Figure 3).

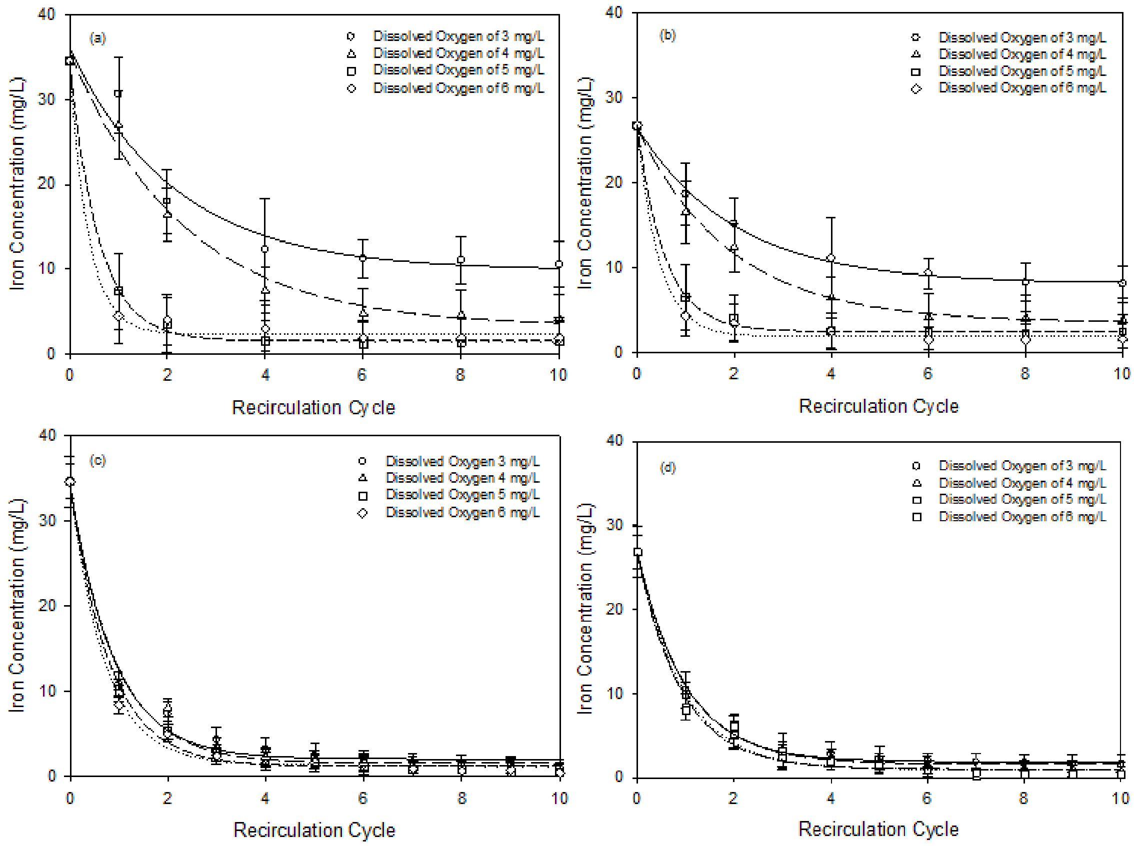

FIGURE 2. IRON REMOVAL FOR LEON COUNTY LANDFILL AND SPRINGHILL LANDFILL LEACHATE AS A FUNCTION OF RECIRCULATION CYCLE IN THE AERATED RECIRCULATION BIOREACTOR. (A) LEON COUNTY LANDFILL LEACHATE AT pH 7.5, (B) SPRINGHILL LANDFILL LEACHATE AT pH 7.5, (C) LEON COUNTY LANDFILL LEACHATE AT pH 8.5, AND (D) SPRINGHILL LANDFILL LEACHATE AT pH 8.5

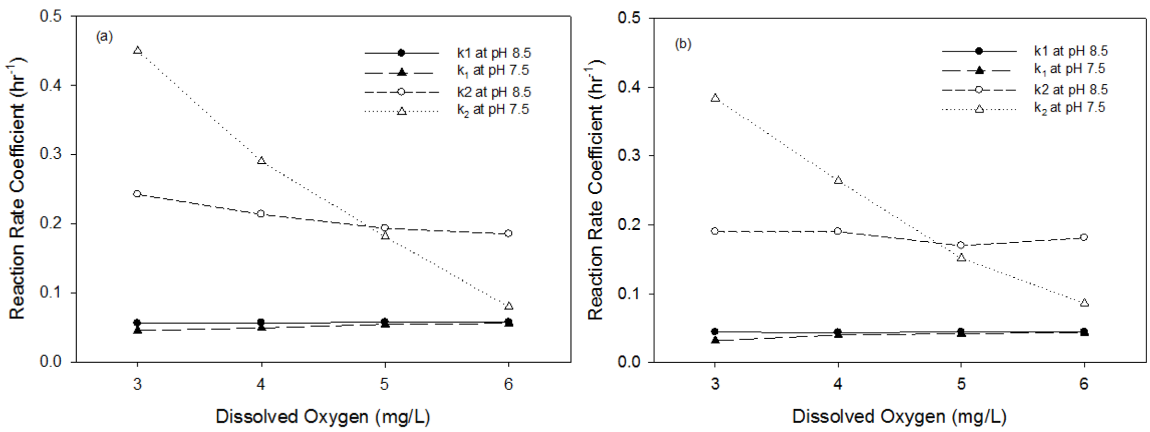

FIGURE 3. SIMULATED IRON REACTION RATE FOR LEON COUNTY LANDFILL AND SPRINGHILL LANDFILL LEACHATE AS A FUNCTION OF DISSOLVED OXYGEN IN THE AERATED RECIRCULATION BIOREACTOR. (A) LEON COUNTY LANDFILL LEACHATE AND (B) SPRINGHILL LANDFILL LEACHATE

Organic removal in the recirculation bioreactor was also studied for landfill leachate collected from the Leon County Landfill and Springhill Landfill. During the recirculation process, organic compounds were released from the solid waste owing to the hydrolysis of organic components besides degradation. Therefore, Equation (1) was also used to simulated organic concentrations to obtain organic release and removal rate coefficients similar to those of iron release and removal. The impact of recirculation cycle on organic release and removal was investigated for 10 recirculation cycles at pH 7.5. After 4 to 5 recirculation cycles, the organic removal became stable. Compared with Leon County Landfill, leachate collected from Springhill Landfill had higher organic contents. The organic removal reached an average of 80% for Springhill Landfill and 70% for Leon County Landfill when the organic removal became stable (Figure 4). Springhill Landfill leachate also had greater organic removal rate coefficient than that of Leon County Landfill leachate, which was in the range of 0.043 hr^{-1} to 0.047 hr^{-1} as compared with 0.10 hr^{-1} to 0.12 hr^{-1}. For both Leon County Landfill and Springhill Landfill leachate, the organic removal rate coefficient increased with the increase of dissolved oxygen, which was more pronounced for Springhill Landfill leachate (Figure 5). In addition, Springhill Landfill had greater organic release rate coefficient than that of Leon County Landfill (0.24 hr^{-1} to 0.08 hr^{-1} as compared with0.12 hr^{-1} to 0.06 hr^{-1}). For both Leon County Landfill and Springhill Landfill, the organic release rate coefficient decreased with the increase of dissolved oxygen. Currently, Leon County Landfill stopped accepting domestic waste and most of the domestic waste from Leon County was deposited in Springhill Landfill. Subsequently, Springhill Landfill contained more easily decomposable organic contents, resulting in higher organic release and removal rate coefficients than those of Leon County Landfill. The BOD/COD ratio was 0.15 for Springhill Landfill leachate as compared with 0.12 for Leon County Landfill leachate. At the most general level, a higher BOD/COD ratio suggested a leachate with higher concentrations of volatile fatty acids that could be easily degraded.

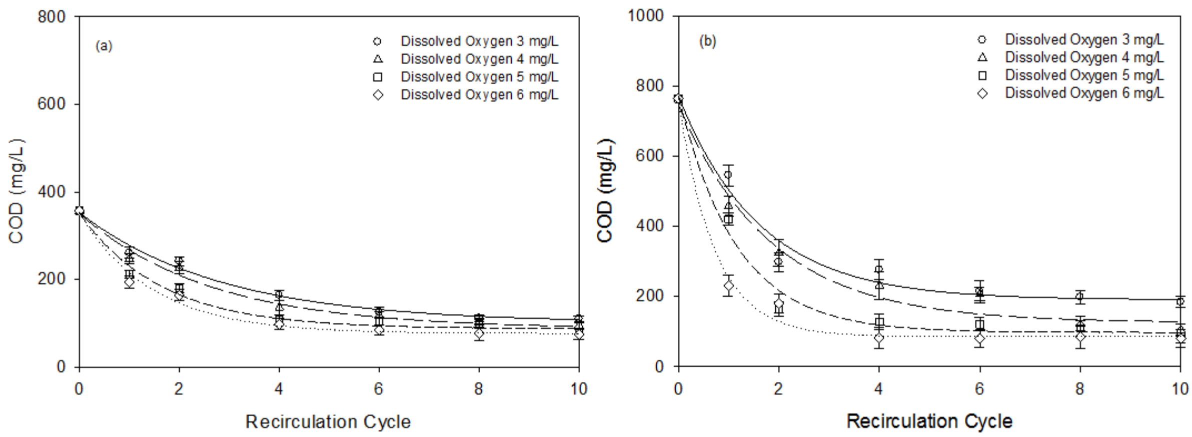

FIGURE 4. ORGANIC REMOVAL FOR (A) LEON COUNTY LANDFILL LEACHATE AND (B) SPRINGHILL LANDFILL LEACHATE AT pH 7.5 AS A FUNCTION OF RECIRCULATION CYCLE IN THE AERATED RECIRCULATION BIOREACTOR

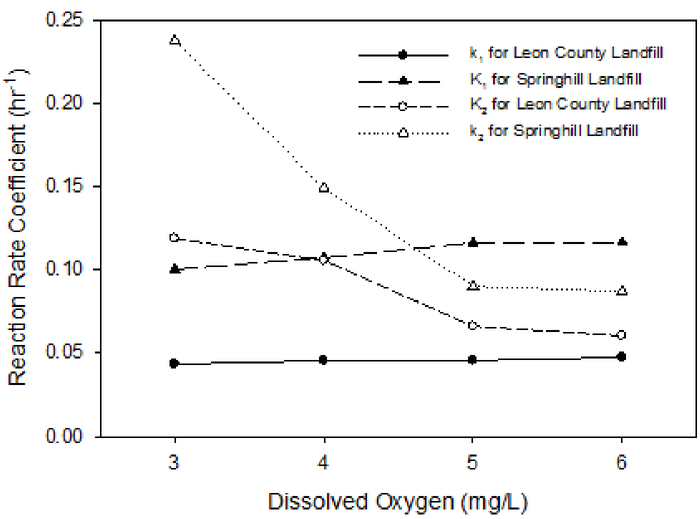

FIGURE 5. SIMULATED ORGANIC REACTION RATE FOR LEON COUNTY LANDFILL AND SPRINGHILL LANDFILL LEACHATE AT pH 7.5 AS A FUNCTION OF DISSOLVED OXYGEN IN THE AERATED RECIRCULATION BIOREACTOR

Pressurized Fiber Biofiltration

After treatment in the aerated recirculation bioreactor, the treated leachate was introduced to the pressurized fiber biofilter. Within the pressurized fiber biofilter, iron removal and organic decomposition were monitored, which were a function of dissolved oxygen and alkalinity. At dissolved oxygen of 1 mg/l and alkalinity of 100 mg/l as $CaCO_3$, the iron removal coefficient was 0.69 hr^{-1} for Leon County landfill leachate and 0.78 hr^{-1} for Springhill Landfill leachate (Figure 6). With the increase of dissolved oxygen, iron removal coefficient increased exponentially, but linearly increased with the increase of alkalinity (Figure 6). Organic removal had similar observations as iron removal. With the increase dissolved oxygen from 1 mg/1 to 3 mg/l, organic removal coefficient increased exponentially from 1.1 hr^{-1} to 2.3 hr^{-1} for Leon County Landfill leachate and 0.98 hr^{-1} to 2.0 hr^{-1} for Springhill Landfill leachate; while with the increase of alkalinity from 100 mg/l as $CaCO_3$ to 800 mg/l as $CaCO_3$, organic removal coefficient increased linearly from 1.35 hr^{-1} to 1.98 hr^{-1} for Leon County landfill leachate and 1.20 hr^{-1} to 1.78 hr^{-1} for Springhill Landfill leachate (Figure 7).

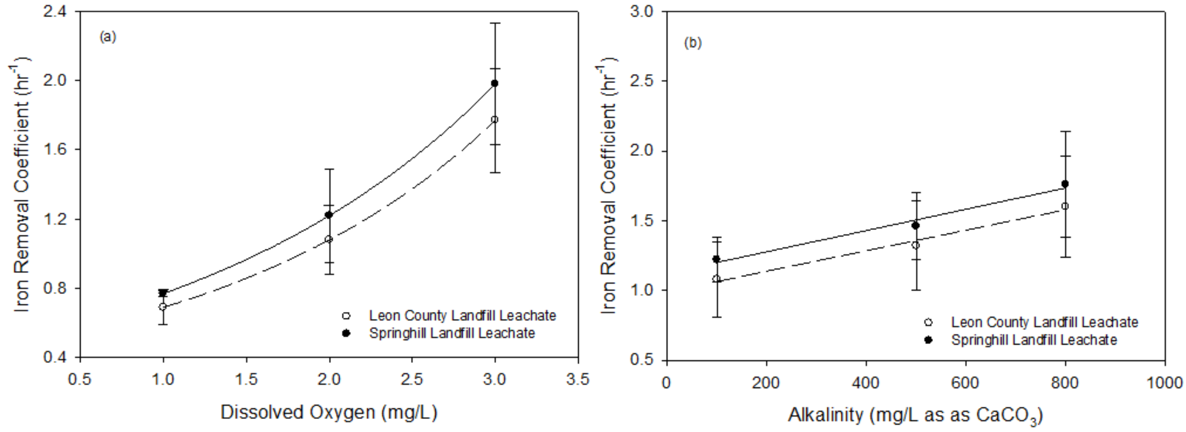

FIGURE 6. SIMULATED IRON REMOVAL COEFFICIENT FOR LEON COUNTY LANDFILL AND SPRINGHILL LANDFILL LEACHATE AS A FUNCTION OF (A) DISSOLVED OXYGEN AND (B) ALKALINITY IN THE PRESSURIZED SUSPENDED FIBER BIOFILTER

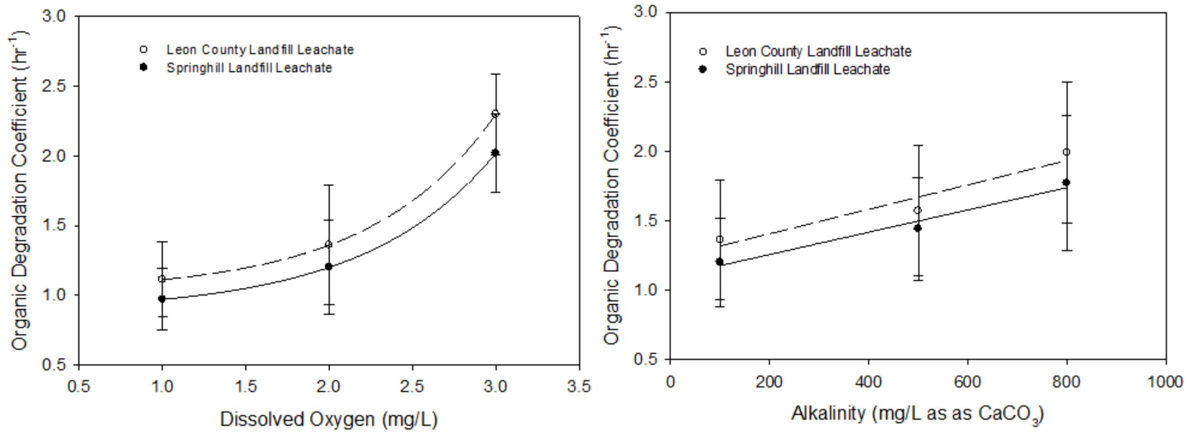

FIGURE 7. SIMULATED ORGANIC REMOVAL COEFFICIENT FOR LEON COUNTY LANDFILL AND SPRINGHILL LANDFILL LEACHATE AS A FUNCTION OF (A) DISSOLVED OXYGEN AND (B) ALKALINITY IN THE PRESSURIZED SUSPENDED FIBER BIOFILTER

Discussion

During landfill operations, it was a common practice to compact waste and cover the waste with a layer of soil each day to reduce odors. Therefore, ferrous iron may be released together with organic decomposition, especially at the bottom section of the landfill when oxygen is depleted. This is attributed to the iron-reducing bacteria that reduce iron oxides to ferrous iron with hydrocarbon-rich landfill leachate serving as the carbon and energy sources as described below:

$$CH_2O + 2Fe_2O_3 + 3H_2O = CO_2 + 4Fe^{2+} + 8OH^- \qquad \text{Equation (7)}$$

Many compounds, in particular easily degradable organic compounds such as volatile fatty acids can be degraded.

Consequently, pH increases as illustrated by Equation (7). Researchers from Florida State University have demonstrated that enrichment cultures of iron-reducing bacteria are capable of conserving energy for growth with the structure Fe (III) bound in smectite clay as the sole electron acceptor (Kostka et al., 2002). Ferrous iron is soluble as a cation, while ferric iron is not. When the leachate was re-circulated to the recirculation bioreactor, oxygen caused Fe(II) to oxidize to Fe(III) and precipitate out of the leachate. Ferrous iron oxidation normally occurs within minutes. After oxidation, ferric iron precipitates in the form of iron hydroxide, $Fe(OH)_3$:

$$Fe^{2+} \xrightarrow{Oxidation} Fe^{3+} \xrightarrow{\Pr ecipitation} Fe(OH)_3 \quad \text{Equation (8)}$$

$Fe(OH)_3$ can be further hydrolyzed to form complex precipitates:

$$[Fe_m(OH)_x]_n^{(3m-x)n+} + yOH^- \rightarrow [Fe_m(OH)_{x+(y/n)}]_n^{[3m-x-(y/n)]n+} \quad \text{Equation (9)}$$

Ferrous iron was mainly removal by aerated recirculation within the top region of the landfill by iron oxidation and subsequent iron hydroxide precipitation within the lower layers of the landfill. The oxidation kinetics of ferrous iron as well as the hydrolysis process is well known to be pH dependent. Change of solution pH usually accompanies with iron hydrolysis because the hydrolysis can consume alkalinity. For landfill leachate with low alkalinity, pH would decrease more obviously with iron hydrolysis. For this research, the pH decrease was not obvious since the hydroxide radical consumption was offset by its production as described in Equation (7). The difference of input and output pH was within tenth of the digit.

Aerated leachate recirculation for the treatment of leachate is a proven technology for organic and ammonia removal in young and intermediate landfills. As a new and promising trend in solid waste management, landfill as a bioreactor such as by means of aerated recirculation has been recognized as one of the lowest cost methods of leachate treatment. There are many advantages associated with treating the landfill as a bioreactor, including the rapid reduction of biodegradable organic compounds and heavy metals in leachate. Typical bioreactor landfill operation is to recycle leachate back through the tip, which has been largely used in the past decade because it was one of the least expensive options available (Renou et al., 2008). Recently, related research has demonstrated benefits of this technique. Significant lowering in methane production and COD was also observed when the recirculated leachate volume was 30% of the initial waste bed volume (Renou et al., 2008). The leachate recycle not only improves the leachate quality, but also shortens the time required for landfill stabilization from several decades to 2 - 3 years (Shen et al., 2001). Although positive effects have been reported on leachate treatment, the recirculation rate may impact further leachate treatment. Especially, high recirculation rates may adversely affect anaerobic degradation of solid waste in the landfill. For instance, leachate recirculation can lead to the inhibition of methanogenesis as it may cause high concentrations of organic acids (pH < 5) which are toxic for the methanogens (Huang et al., 2008). Furthermore, if the volume of leachate recirculated is very high, problems such as saturation, ponding and acidic conditions may occur (San and Onay, 2001).

Biofilters are different from conventional gravity filters and can not only filter suspended solid, but also increase the degradation of organic matter and iron removal using the fixed film biomass. Inside the biofilter, iron fixation bacteria fixed iron to the fiber using the energy obtained from iron oxidation. On the other hand, hydrocarbons provided a carbon source for heterotrophic bacteria. Both of these processed consumed oxygen. Therefore, air was continuously provided. Thermodynamically, organic decomposition was more favorable in the system than ferrous iron oxidation, which was consistent with the observation of this research. The thus calculated energy release of organics was -28.7 kcal, -26.4 kcal and -25.3 kcal for carbohydrates, proteins and fat as compared with -0.89 kcal for ferrous iron oxidation based on the Gibbs free energy of related half reactions (Table 1). Therefore, organic decomposition was favorable in the fiber bioreactor because of the higher energy production. The iron oxidation process was limited by the energy production. In addition, the iron oxidizing bacteria grew slower as compared with heterotrophic strains owing to the smaller yield coefficient. Biofilm thickness was proportional to the organic content in the leachate. Owing to the high organic content in Springhill Landfill leachate, thicker biofilm was observed. Biofilm activity was not proportional to the quantity of fixed biomass, but increased with the depth of biofilter. The biofilm on the top layer was thinner than that of the bottom section of the biofilter. This was verified by the fiber observations after treatment. The massive biomass ultimately resulted in the progressive clogging of the biofilter, which must then be washed clean. The biofilter clogging was more pronounced in the bottom layer.

Iron fixation in the pressurized suspended fiber biofilter was evidenced by observations of the fiber under a scanning electronic microscope. Crystal iron was found to be fixed to the fiber by iron oxidizing bacteria (Figure 8).

TABLE 1. GIBBS FREE ENERGY OF HALF REACTIONS

Reactions for Organic Compounds	ΔG^0 (kCal/e^- eq)
Carbohydrate: 1/24 $C_6H_{12}O_6$ + 1/4 H_2O = 1/4 CO_2 + H^+ + e^-	-10.0
Protein: 1/66 $C_{16}H_{22}O_5N_4$ + 27/66 H_2O = 8/33 CO_2 + 2/33 NH_4^+ + 31/33 H^+ + e^-	-7.7
Grease, Fat and Oil: 1/46 $C_8H_{16}O$ + 1/4 H_2O = 1/4 CO_2 + H^+ + e^-	-6.6
Reactions for Iron Oxidation	
Fe^{3+} + e^- = Fe^{2+}	74.3
Reactions for Electron Acceptors	
1/4 O_2 + H^+ + e^- = 1/2 H_2O	18.7

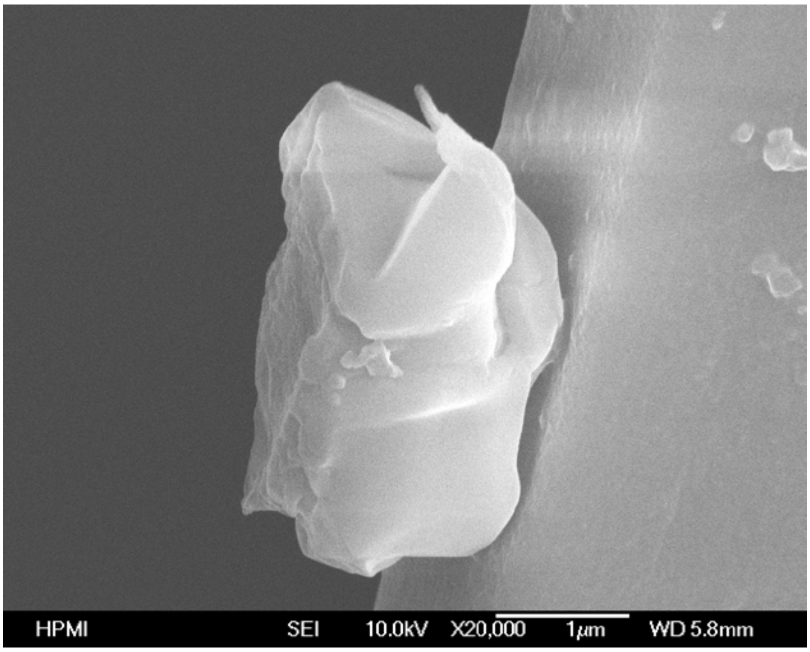

FIGURE 8. CRYSTAL STRUCTURE OF IRON DEPOSITED ON POLYPROPYLENE FIBER OBSERVED UNDER A SCANNING ELECTRONIC MICROSCOPE

Biofiltration has a promising potential for landfill leachate treatment. Several low-cost filter materials have been investigated, which could be included in the systems based on filter-bed techniques (Cohen, 2001). For organic and heavy metal removal from landfill leachate, three filter media, i.e., sand, blast-furnace slag (BFS) and polonite have been investigated, which is based on the determination of contaminant saturation potential in a long-term column study (Cohen, 2001). This method provides a good indicator of capacity of the filter materials to retain elements, which could serve as a parameter for estimating the lifetime of full-scale systems. Pressurized biofilters can further enhance contaminant removal from landfill leachate, which have been practiced for organic and inorganic removal, radiological removal, iron and manganese control, water softening, and pH adjustment, etc. Typically, pressurized biofilters are used for the removal of iron and manganese as well as arsenic (Jessen et al., 2005). From this research, it was demonstrated that pressurized suspended fiber biofilters could be used for the treatment of landfill leachate with high organic and iron contents. It was proven that pressurized suspended fiber biofilters could achieve the treatment goals to different extents under different dissolved oxygen and alkalinity conditions.

Conclusions

From this research, it was demonstrated that landfill leachate could be treated by aerated recirculation and pressurized suspended fiber biofiltration to remove organics and iron. This treatment process started with aerated

recirculation, which also improved the leachate quality and shortened the time required for landfill stabilization besides organic and iron removal. However, during aerated recirculation, in addition to organic and iron removal, organic and iron were also released from the solid waste, especially at the bottom section of the landfill when oxygen was depleted. The pressurized suspended fiber biofilter was designed and operated under pressurized aeration to achieve biological contact oxidation, which can remov organic compounds and iron more efficiently than conventional biological methods. Iron removal coefficient in the pressurized suspended fiber biofilter increased exponentially with the increase of dissolved oxygen, but linearly increased with the increase of alkalinity. Organic removal had similar observations. Thermodynamically, organic decomposition was favorable in the pressurized suspended fiber biofilter because of the high energy production; while iron oxidation process was limited owing to the low energy production. In addition, iron oxidizing bacteria grew slower as compared with heterotrophic strains owing to the smaller yield coefficient. Within the pressurized suspended fiber biofilter, biofilm activity was not proportional to the quantity of fixed biomass, but increased with the depth of biofilter. Iron fixation in the pressurized suspended fiber biofilter was evidenced by observations of the fiber under a scanning electronic microscope and crystal iron was found to be fixed to the fiber by iron oxidizing bacteria.

ACKNOWLEDGEMENT

The work was support by Hinkley Center for Solid and Hazardous Waste Management through Grant No. UF-EIES-1332025-FSU to Florida State University.

REFERENCES

[1] Bilgili, Sinan M., Demir, Ahmet, and Ozkaya, Bestamin. "Influence of Leachate Recirculation on Aerobic and Anaerobic Decomposition of Solid Wastes". Journal of Hazardous Materials 143 (2007): 177-183.

[2] Chaiprasert, Pawinee, Suvajittanont, Worakrit, Suraraksa, Benjaphon, Tanticharoen, Morakot, and Bhumiratana, Sakarindr. (2003). "Nylon Fibers as Supporting Media in Anaerobic Hybrid Reactors: It's Effects on System's Performance and Microbial Distribution". Water Research 37 (2003): 4605-4612.

[3] Coban, Asli, Demir, Goksel, Okten, Hatice Eser, Ozcan, Kurtulus H., Yaman, Cevat, and Yildiz, Senol. "Advanced Treatment of Leachate by Using Aerobic/Anoxic MBR System Followed by a Nanofiltration Process. A Case Study in Istanbul Komurcuoda Leachate Treatment Plant". Environment Protection Engineering 38 (2012): 57-64.

[4] Cohen, Yariv. "Biofiltration — the Treatment of Fluids by Microorganisms Immobilized into the Filter Bedding Material: A Review". Bioresource Technology 77 (2001): 257-74.

[5] Ellis, K. V., Mortimer, G. H., and Berktay, A. "Biological Wastewater Treatment under the Influence of Pressure". Journal of the Institution of Water and Environmental Management 6 (1992): 468-474.

[6] Ferraz, F. M., Povinelli, J., Pozzi, E., Vieira, E. M., and Trofino, J. C. "Co-Treatment of Landfill Leachate and Domestic Wastewater Using a Submerged Aerobic Biofilter". Journal of Environmental Management 141 (2014): 9-15.

[7] Huang, Qifei, Yang, Yufei, Pang, Xiangrui, and Wang, Qi. "Evolution on Qualities of Leachate and Landfill Gas in the Semi-Aerobic Landfill". Journal of Environmental Science (China) 20 (2008): 499-504.

[8] Jessen, Soren, Larsen, Flemming, Koch, Christian Bender, and Arvin, Eeik. "Sorption and Desorption of Arsenic to Ferrihydrite in a Sand Filter". Environmental Science and Technology 39 (2005): 8045-51.

[9] Kostka, Joel E., Dalton, Dave D., Skelton, Hayley, Dollhopf, Sherry, and Stucki, Joseph W. "Growth of Iron(III)-Reducing Bacteria on Clay Minerals as the Sole Electron Acceptor and Comparison of Growth Yields on a Variety of Oxidized Iron Forms". Applied and Environmental Microbiology 68 (2002): 6256-6262.

[10] Lee, J. J., Cha, J. H., Ben Aim, R., Han, K. B., and Kim, C. W. "Fiber Filter as an Alternative to the Process of Flocculation-Sedimentation for Water Treatment". Desalination 231 (2008): 323-331.

[11] Lovley, Derek R., and Phillips, Elizabeth J. P. "Novel Mode of Microbial Energy Metabolism: Organic Carbon Oxidation Coupled to Dissimilatory Reduction of Iron or Manganese". Applied and Environmental Microbiology 54 (1988): 1472-1480.

[12] McCarty, Perry L. “Stoichiometry of Biological Reactions”. Progress in Water Technology 7 (1975): 157-172.

[13] Nikolaou, A., Giannis, A., and Gidarakos, E. “Comparative Studies of Aerobic and Anaerobic Treatment of MSW Organic Fraction in Landfill Bioreactors”. Environmental Technology 31 (2010): 1381-1389.

[14] Ragle, Nancy, Kissel, John, Ongerth, Jerry E., and Dewalle, Foppe B. “Composition and Variability of Leachate from Recent and Aged Areas within a Municipal Landfill”. Water Environment Research 67 (1995): 238-243.

[15] Reinhart, Debra R., and AlYousfi, Basel A. “The Impact of Leachate Recirculation on Municipal Solid Waste Landfill Operating Characteristics”. Waste Management and Research 14 (1996): 337-346.

[16] Renou, S., Givaudan, J. G., Poulain, S., Dirassouyan, F., and Moulin, P. Landfill leachate treatment: Review and opportunity. Journal of Hazardous Materials 150 (2008): 468-493.

[17] Roden, E. E., and Zachara, J. M. “Microbial Reduction of Crystalline Iron(III) Oxides: Influence of Oxide Surface Area and Potential for Cell Growth”. Environmental Science and Technology 30 (1996): 1618-1628.

[18] San, Irem, and Onay, Turgut T. “Impact of Various Leachate Recirculation Regimes on Municipal Solid Waste Degradation”. Journal of Hazardous Materials 87 (2001): 259-71.

[19] Shen, Dongsheng, He, Ruo, Ren, Guoping, Traore, I., and Feng, Xiaoshen. “Effect of Leachate Recycle and Inoculation on Microbial Characteristics of Municipal Refuse in Landfill Bioreactors”. Journal of Environmental Science (China) 13 (2001): 508-13.

[20] Williams, Mitchell D., Subramanian, Pawan K., and Chen, Gang. “Soil and Microbial Characterization and Microbial Mediated Iron Release nearby Landfills in Northwest Florida, U.S." International Journal of Environmental Waste Management 10 (2012): 56-69.

[21] Wintheiser, P. “Leachate Recirculation: A Review of Operating Experience at Municipal Solid Waste Landfills throughout the United States”. Proceedings from Swana’s 3rd Annual Landfill Symposium (1998): 59-62.

[22] Yang, Yufei, Yue, Bo, Yang, Yu, and Huang, Qifei. “Influence of Semi-Aerobic and Anaerobic Landfill Operation with Leachate Recirculation on Stabilization Processes”. Waste Management and Research 30 (2012): 255-265.